

WHAT IS CLAIMED IS:

1. A coating liquid for forming an amorphous silica-based coating film with a low dielectric constant having a high film strength and excellent hydrophobic property and capable of ensuring smoothness of a surface coated therewith,

wherein the coating liquid contains a silicon compound obtained by hydrolyzing tetraalkyl ortho silicate (TAOS) and alkoxysilane (AS) expressed by the following general formula (I) in the presence of tetraalkyl ammonium hydroxide (TAAOH):



wherein X indicates a hydrogen atom, a fluorine atom, or an alkyl group, a fluorine-substituted alkyl group, an aryl group or a vinyl group each having 1 to 8 carbon atoms; R indicates a hydrogen atom, or an alkyl group, an aryl group or a vinyl group each having 1 to 8 carbon atoms; and n is an integral number from 0 to 3.

2. A coating liquid for forming an amorphous silica-based coating film with a low dielectric constant having a high film strength and excellent hydrophobic property and capable of ensuring smoothness of a surface coated therewith,

wherein the coating liquid contains a silicon compound obtained by hydrolyzing or partially hydrolyzing tetraalkyl ortho silicate (TAOS) in the presence of tetraalkyl ammonium hydroxide (TAAOH), mixing a reaction product with the alkoxysilane (AS) expressed by the general formula (I) above or a hydrolysate or a partial hydrolysate thereof, and further hydrolyzing all or a portion of a mixture according to the necessity.

3. The coating liquid for forming a silica-based coating film with a low dielectric constant according to any of claims 1 to 2, wherein said tetraalkyl ortho silicate (TAOS)

is tetraethyl ortho silicate (TEOS), tetramethyl ortho silicate (TMOS) or a mixture thereof.

4. The coating liquid for forming a silica-based coating film with a low dielectric constant according to any of claims 1 to 3, wherein said alkoxysilane (AS) is methyltrimethoxy silane (MTMS), methyltriethoxy silane (MTES) or a mixture thereof.

5. The coating liquid for forming a silica-based coating film with a low dielectric constant according to any of claims 1 to 4, wherein said tetraalkyl ammonium hydroxide (TAAOH) is tetrapropyl ammonium hydroxide (TPAOH), tetrabutyl ammonium hydroxide (TBAOH) or a mixture thereof.

6. The coating liquid for forming a silica-based coating film with a low dielectric constant according to any of claims 1 to 5, wherein a content of impurities comprising compounds of alkali metal elements such as sodium (Na) and potassium (K) in said tetraalkyl ammonium hydroxide (TAAOH) is 50 ppb by weight or below on respective element bases.

7. The coating liquid for forming a silica-based coating film with a low dielectric constant according to any of claims 1 to 6, wherein a content of impurities comprising compounds of halogen group elements such as bromine (Br) and chlorine (Cl) in said tetraalkyl ammonium hydroxide (TAAOH) is 1 ppm by weight or less on respective element bases.

8. The coating liquid for forming a silica-based coating film with a low dielectric

constant according to any of claims 1 to 7, wherein a molar ratio (TAOS/AS) of said tetraalkyl ortho silicate (TAOS) and said alkoxysilane (AS) is in a range from 6/4 to 2/8 in terms of SiO₂.

5 9. The coating liquid for forming a silica-based coating film with a low dielectric constant according to any of claims 1 to 8, wherein a molar ratio (TAAOH/(TAOS+AS)) of said tetraalkyl ammonium hydroxide (TAAOH) and the components for forming the silica-based coating film (TAOS+AS) is in a range from 1/10 to 7/10 in terms of SiO₂.

10 10. The coating liquid for forming a silica-based coating film with a low dielectric constant according to any of claims 1 to 9, wherein the coating liquid contains a silicon compound as a hydrolysate of said tetraalkyl ortho silicate (TAOS) and said alkoxysilane (AS) by 2 to 40% by weight.

15 11. A method of preparing a coating liquid for forming an amorphous silica-based coating film with a low dielectric constant having a high film strength and excellent hydrophobic property and ensuring smoothness of a surface coated therewith comprising the steps of:

20 (i) mixing tetraalkyl ortho silicate (TAOS) and alkoxysilane (AS) expressed by the following general formula (I) with an organic solvent and agitating the mixture at a temperature in a range from 10 to 30 °C until the components are fully mixed with each other at a rotating speed of 100 to 200 rpm,

(ii) adding an aqueous solution of tetraalkyl ammonium hydroxide (TAAOH)
25 into the mixture solution under agitation over 5 to 20 minutes, and further agitating the

mixture solution for 30 to 90 minutes at a temperature in a range from 10 to 30 °C at a rotating speed of 100 to 200 rpm, and then

(iii) heating the mixture solution at a temperature in a range from 30 to 80 °C and further agitating the mixture solution for 1 to 72 hours at a rotating speed of 100 to 200 rpm keeping the temperature at the same level to prepare a liquid composition containing a silicon compound which is said hydrolysate of the tetraalkyl ortho silicate (TAOS) and the alkoxysilane (AS):



wherein X indicates a hydrogen atom, fluorine atom, or an alkyl group, a fluorine-substituted alkyl group, an aryl group or a vinyl group each having 1 to 8 carbon atoms; R indicates a hydrogen atom, or an alkyl group, an aryl group or a vinyl group each having 1 to 8 carbon atoms; and n is an integral number from 0 to 3.

12. The method of preparing a coating liquid for forming a coating film according to claim 11 comprising the step of:

in place of the adding method according to claim 11, slowly adding the mixture solution comprising the tetraalkyl ortho silicate (TAOS), the alkoxysilane (AS) and the organic solvent prepared in said step (i) into the aqueous solution of the tetraalkyl ammonium hydroxide (TAAOH) in said step (ii) over 30 to 90 minutes under the same conditions as those in claim 11.

13. A method of preparing a coating liquid for forming an amorphous silica-based coating film with a low dielectric constant having a high film strength and excellent hydrophobic property and capable of ensuring smoothness of a surface coated therewith comprising the steps of:

(i) mixing tetraalkyl ortho silicate (TAOS) with an organic solvent and agitating the mixture at a temperature in a range from 10 to 30 °C until the components are fully mixed with each other at a rotating speed of 100 to 200 rpm;

(ii) adding an aqueous solution of tetraalkyl ammonium hydroxide (TAAOH) into the mixture solution under agitation over 5 to 20 minutes, and further agitating the mixture solution for 30 to 90 minutes at a temperature in a range from 10 to 30 °C at a rotating speed of 100 to 200 rpm;

(iii) heating the mixture solution to a temperature in a range from 30 to 80 °C and agitating the mixture solution keeping the temperature for 0.5 to 72 hours at a rotating speed of 100 to 200 rpm to prepare a mixture solution containing a hydrolysate and/or a partial hydrolysate of the tetraalkyl ortho silicate (TAOS);

(iv) further mixing alkoxysilane (AS) expressed by the general formula (I) above or a mixture thereof with an organic solvent in the mixture solution obtained in said step (iii), and agitating the resultant mixture solution at a temperature in a range from 10 to 30 °C until the components are fully mixed with each other at a rotating speed of 100 to 200 rpm;

(v) adding an aqueous solution of tetraalkyl ammonium hydroxide (TAAOH) into the mixture solution under agitation over 5 to 20 minutes, and further agitating the resultant mixture solution at a temperature from 10 to 30 °C for 30 to 90 minutes at a rotating speed of 100 to 200 rpm; and then

(vi) heating the mixture solution obtained in said step (v) to a temperature in a range from 30 to 80 °C and agitating the mixture solution keeping the temperature for 10 to 30 hours at a rotating speed of 100 to 200 rpm to prepare a liquid composition containing a silicon compound which is a hydrolysate of the tetraalkyl ortho silicate (TAOS) and the alkoxysilane (AS).

14. The method of preparing a coating liquid for forming a coating film according to claim 13 comprising the step of:

in place of the adding method according to claim 13, slowly adding the mixture
5 solution comprising the tetraalkyl ortho silicate (TAOS) and the organic solvent prepared in said step (i) into the aqueous solution of tetraalkyl ammonium hydroxide (TAAOH) in said step (ii) over 30 to 90 minutes under the same conditions as those in claim 13.

15. A method of preparing a coating liquid for forming an amorphous silica-based
10 coating film with a low dielectric constant having a high film strength and excellent hydrophobic property and ensuring smoothness of a surface coated therewith comprising the steps of:

(i) mixing tetraalkyl ortho silicate (TAOS) with an organic solvent and
agitating the mixture at a temperature in a range from 10 to 30 °C at a rotating speed of
15 100 to 200 rpm until the components are fully mixed with each other;

(ii) adding an aqueous solution of tetraalkyl ammonium hydroxide (TAAOH)
into the mixture solution under agitation over 5 to 20 minutes, and further agitating the
mixture solution for 30 to 90 minutes at a temperature in a range from 10 to 30 °C at a
rotating speed of 100 to 200 rpm;

20 (iii) heating the mixture solution to a temperature in a range from 30 to 80 °C and agitating the mixture solution keeping the temperature for 0.5 to 72 hours at a rotating speed of 100 to 200 rpm to prepare a mixture solution containing a hydrolysate and/or a partial hydrolysate of said tetraalkyl ortho silicate (TAOS);

(iv) further mixing alkoxysilane (AS) expressed by the general formula (I)
25 above and an organic solvent in the mixture solution above, and agitating the resultant

solution at a temperature in a range from 10 to 30 °C and at a rotating speed of 100 to 200 rpm until the components are fully mixed with each other;

(v) adding an aqueous solution of tetraalkyl ammonium hydroxide (TAAOH) into the mixture solution above under agitation over 5 to 20 minutes, and then agitating the resultant mixture solution at a temperature from 10 to 30 °C for 30 to 90 minutes at a rotating speed of 100 to 200 rpm;

(vi) then heating the mixture solution to a temperature in a range from 30 to 80 °C, and agitating the mixture solution keeping the temperature for 0.5 to 72 hours at a rotating speed of 100 to 200 rpm to prepare a mixture solution containing a hydrolysate and/or a partial hydrolysate of said alkoxysilane (AS);

(vii) mixing the mixture solution obtained in said step (iii) and that obtained in said step (vi) and agitating the resultant mixture solution at a temperature in a range from 10 to 30 °C and at a rotating speed of 100 to 200 rpm until the components are fully mixed with each other; and

(viii) further heating the solution obtained in said step (vii) to a temperature from 30 to 80 °C according to the necessity and agitating the solution keeping the temperature for 10 to 30 hours at a rotating speed of 100 to 200 rpm to prepare a liquid composition containing a silicon compound which is a hydrolysate of the tetraalkyl ortho silicate (TAOS) and the alkoxysilane (AS).

16. The method of preparing a coating liquid for forming a coating film according to claim 15 comprising the steps of:

in place of the adding method according to claim 15, slowly adding the mixture solution comprising tetraalkyl ortho silicate (TAOS) and organic solvent prepared in said step (i) into an aqueous solution of the tetraalkyl ammonium hydroxide (TAAOH) in said

step (ii) over 30 to 90 minutes under the same conditions as those in claim 15, and also slowly adding the mixture solution comprising alkoxysilane (AS) and organic solvent prepared in said step (iv) over 30 to 90 minutes into an aqueous solution of the tetraalkyl ammonium hydroxide (TAAOH) in said step (v) under the same conditions as those in claim 15.

17. The method of preparing a coating liquid for forming a silica-based coating film with a low dielectric constant according to any of claims 11 to 16, wherein said tetraalkyl ortho silicate (TAOS) is tetraethyl ortho silicate (TEOS), tetramethyl ortho silicate (TMOS) or a mixture thereof.

18. The method of preparing a coating liquid for forming a silica-based coating film with a low dielectric constant according to any of claims 11 to 17, wherein said alkoxysilane (AS) is methytrimethoxy silane (MTMS), methyltriethoxy silane (MTES) or a mixture thereof.

19. The method of preparing a coating liquid for forming a silica-based coating film with a low dielectric constant according to any of claims 11 to 18, wherein said tetraalkyl ortho silicate (TAOS) and said alkoxysilane (AS) are mixed with each other at a molar ratio (TAOS/AS) in a range from 6/4 to 2/8 in terms of SiO_2 .

20. The method of preparing a coating liquid for forming a silica-based coating film with a low dielectric constant according to any of claims 11 to 19, wherein said tetraalkyl ammonium hydroxide (TAAOH) is tetrapropyl ammonium hydroxide (TPAOH), tetrabutyl ammonium hydroxide (TBAOH) or a mixture thereof.

21. The method of preparing a coating liquid for forming a silica-based coating film with a low dielectric constant according to any of claims 11 to 20, wherein said aqueous solution of tetraalkyl ammonium hydroxide (TAAOH) is an aqueous solution containing
5 tetraalkyl ammonium hydroxide (TAAOH) by 5 to 40% by weight.

22. The method of preparing a coating liquid for forming a silica-based coating film with a low dielectric constant according to any of claims 11 to 21, wherein said aqueous solution of tetraalkyl ammonium hydroxide (TAAOH) is added to components for forming
10 a silica-based coating film (TAOS+AS) at a molar ratio (TAAOH/(TAOS+AS)) of 1/10 to 7/10 in terms of SiO_2 .

23. The method of preparing a coating liquid for forming a silica-based coating film with a low dielectric constant according to any of claims 11 to 22, wherein a content of
15 impurities comprising compounds of alkali metal elements such as sodium (Na) and potassium (K) contained in said aqueous solution of tetraalkyl ammonium hydroxide (TAAOH) is 50 ppb by weight or less on each element basis.

24. The method of preparing a coating liquid for forming a silica-based coating film
20 with a low dielectric constant according to any of claims 11 to 23, wherein a content of impurities comprising compounds of halogen group elements such as bromine (Br) and chlorine (Cl) is 1 ppm by weight or less on each element basis.

25. The method of preparing a coating liquid for forming a silica-based coating film
25 with a low dielectric constant according to any of claims 20 to 24, wherein said aqueous

solution of tetraalkyl ammonium hydroxide (TAAOH) is obtained by subjecting a commercially available aqueous solution of tetraalkyl ammonium hydroxide to a cation exchange resin treatment and also to an anion exchange resin treatment to substantially remove impurities such as compounds of alkali metal elements such as sodium (Na) and potassium (K) as well as those of halogen group elements such as bromine (Br) and chlorine (Cl) contained therein for high-level purification.

26. The method of preparing a coating liquid for forming a silica-based coating film with a low dielectric constant according to any of claims 11 to 25, wherein said organic solvent is alcohol such as ethanol.

27. The method of preparing a coating liquid for forming a silica-based coating film with a low dielectric constant according to any of claims 11 to 26, wherein the organic solvent contained in the liquid composition obtained by the method according to any of claims 11 to 26 is subjected to a process for solvent-substituting with an organic solvent selected among propylene glycol monopropyl ether (PGP), propylene glycol monomethyl ether (PGME), propylene glycol monoethyl ether acetate (PGMEA) and the like employing a rotary evaporator.

28. The method of preparing a coating liquid for forming a silica-based coating film with a low dielectric constant according to any of claims 11 to 27, wherein said liquid composition obtained is adjusted so that said silicon compound which is the hydrolysate of tetraalkyl ortho silicate (TAOS) and alkoxysilane (AS) is contained in the liquid composition by 2 to 40 % by weight.